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We have observed intense band emissions between 340 nm and 344 nm from electron-beam excited mixtures of high pressure argon gas and iodine vapor. This emission appears to be from molecular iodine. Using the 357.6 nm band from an Ar/N_2 mixture as a calibration, we have measured the fluorescence yield in this wavelength region to be 70 ± 24%. From this, we computed an over-all fluorescence energy efficiency of 13 ± 4%. Based on these observations, we suggest the feasibility of an efficient high energy electron-beam pumped argon-iodine laser.

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The gain of such an argon-iodine laser should be comparable to that of the Ar/N_2 second positive laser.

SUMMARY

The purpose of this ARPA-supported program is to develop an understanding of the molecular kinetics pertinent to new high efficiency, high power lasers. We are studying high pressure gases that are initially excited by intense bursts of electrons. This initial pumping energy, which is primarily deposited by creating atomic ions, rapidly collects in the lowest molecular excited state with an overall efficiency of the rare gases near 50%.

Rare gas dimers for which the ground level is repulsive, in particular Xe_2, Kr_2, and Ar_2, have already demonstrated laser action but with disappointing efficiencies. This problem, coupled with the relatively high gain (implying low energy storage capacity) and with vacuum ultraviolet wavelengths, has led us to consider ways of transferring the energy deposited in the rare gas to other gas molecules to improve total efficiency, shift the wavelength to the near ultraviolet or visible, and improve the energy storage capacity.

This report summarizes the studies we have recently made on e-beam pumped mixtures of argon and iodine. We have observed very high fluorescence yield in a band near 340 nm from such mixtures. Since the potential curves of the iodine molecular state responsible for this emission are offset with respect to internuclear separation, we anticipated that this medium would have good prospect as a laser candidate.

This manuscript was submitted in April to Applied Physics Letters, and will be published in the September 15 edition of that Journal. Copies were also sent to several laboratories where e-beam pumped laser research is carried out. Subsequent to our suggestion, at least two of these laboratories have observed lasing on this transition in molecular iodine.

J. J. Ewing from AVCO-Everett reported that he observed lasing not only from molecular iodine mixed with argon, but also from mixtures of argon gas and iodine-bearing compounds such as CF_3I and HI. M. Bhaumik reported to us that the laser laboratory at Northrop has observed lasing from CF_3I and that the computed efficiency is rather promising. We are currently carring out more detailed investigations of the kinetics of these systems in order to understand the mechanisms for populating the upper levels and removing the lower level of this transition.

During the last reporting period, in addition to the discovery of high pressure argon/iodine mixtures as a potential high efficiency laser medium, we have also continued our studies of e-beam pumped mixtures of mercury and xenon. A detailed account of these investigations will appear shortly in the Final Technical Report for this contract.

THE POSSIBILITY OF AN EFFICIENT TUNABLE MOLECULAR IODINE LASER NEAR 340 nm*

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ABSTRACT

We have observed intense band emissions between 340 nm and 344 nm from electron-beam excited mixtures of high pressure argon gas and iodine vapor. This emission appears to be from molecular iodine. Using the 357.6 nm band from an Ar/N_2 mixture as a calibration, we have measured the fluorescence yield in this wavelength region to be 70 $\pm$ 24%. From this we computed an over-all fluorescence energy efficiency of 13 $\pm$ 4%. Based on these observations, we suggest the feasibility of an efficient high energy electron-beam pumped argon-iodine laser. The gain of such an argon-iodine laser should be comparable to that of the Ar/N_2 second positive laser.

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The search for new ultraviolet lasers that are highly efficient has been particularly stimulated by the requirements of isotope separation programs, laser-induced thermonuclear fusion schemes, and military applications. Electron-beam pumping of high pressure noble gases is known to be very efficient.[1-3] This has motivated us to investigate promising laser systems that utilize collisional energy transfer from e-beam excited noble gases to create population inversions between electronic states of acceptor molecules.[4] These molecules can be selected to have lasing transitions with wavelengths suitable for various applications. For example, we have made detailed kinetic studies of the argon-nitrogen[3-5] and xenon-oxygen[6,7] system, in which efficient visible and u.v. laser action has subsequently been demonstrated.[8-11] In this letter we suggest that an argon-iodine mixture is a particularly promising candidate for a laser of this type.

The experimental apparatus and procedures used in this study have been described elsewhere.[3] Briefly, argon gas at a pressure between one-half and four atmospheres and low pressure iodine vapor (99.95% purity) were contained in a graphite coated stainless steel vessel; a rotating vane in the vessel promoted rapid mixing. The iodine vapor pressure was controlled by adjusting the temperature of the vessel and the iodine reservoir. Electrons from a Febetron 706 pulsed accelerator entered the reaction chamber through a thin metal foil in the wall. Light emitted from the cylindrical reaction region passed through sapphire windows into a spectrometer (McPherson Model 216.5) was analyzed in second order

(resolution 0.8 Å), and recorded on Polaroid film. The time behavior of the emission was recorded on photographs of oscilloscope traces produced from the output of a fast photomultiplier (1P28 or R666). Direct comparisons of emission intensities with the calibrated N_2 ($C^3\Pi_u \rightarrow B^3\Pi_g$) second positive band were made by flushing the Ar-I_2 mixture from the reaction chamber and refilling with Ar + 5% N_2 at the same total pressure.

The brightest emission feature observed in the 250 nm to 900 nm region was a banded structure (shown in Figure 1) extending from approximately 300.0 to 345.0 nm with most of the intensity between 340.0 and 343.0 nm and a general decrease in intensity toward the blue. The series displayed obvious regularity, with local red degraded maxima occurring every 0.95 to 1.05 nm down to 300.0 nm.[12] A weaker band, 2.0 nm wide, was recorded near 287.0 nm, and other bands were observed near 430.0 nm and 450.0 nm with peak intensities at least a factor of 100 less than the 340 nm band.

The exponential rise and fall times of all these emissions were approximately independent of wavelength. For example, with two atmospheres of argon and room temperature iodine ($\sim \frac{1}{4}$ torr), the rise time is 105 ns, and the decay time is 225 ns. Increased iodine pressure yielded increased peak intensity, reduced decay time, and only slightly reduced time-integrated intensity. Detailed studies of the kinetic effects leading to such behavior are underway and will be reported later.

The bright emissions near 340.0 nm have been previously observed with various excitation sources, and both with and without the presence

of an argon buffer.[13,14] While Mulliken[15] tentatively suggested that both D-X and $^3\Pi_{2g} - {}^3\Pi_{2u}$ transitions may be present in this band (see Fig. 2), recent studies have indicated that the spectroscopic assignments may be more complicated.[13,16] It is clear, however, that these emissions arise from electronic transitions between upper ionic and lower covalent states of I_2. Moreover, the potential minima for these states are at substantially different internuclear separations.

Our present knowledge of the kinetic mechanism that populates these upper ionic states of I_2 is incomplete. This excitation process may involve collisional or radiative transfer of energy from the excited argon atoms and excimers to the molecular iodine. Since the argon excimer energy exceeds the ionization potential of I_2, ionization or dissociation (perhaps to I^+ or I^-) and subsequent recombination may be involved. Such processes could be highly selective in the production of the ionic states of the I_2 molecule and thus efficiently create the population inversion needed for lasing.

Tellinghuisen[16] recently considered the possibility of a photon-pumped laser operating between high lying vibrational levels of the excited D state of I_2 and high vibrational levels of the ground (X) state. Electron-beam excitation of high pressure argon/iodine mixtures offers obvious advantages, in addition to the high energy conversion efficiency of e-beam pumped noble gases. The excited I_2 population will be rapidly

relaxed into just a few vibrational levels by collisions, and bottlenecking will be prevented by rapid vibrational relaxation of the lower level.

At 342.5 nm the peak light flux from 2 atm. argon and $\frac{1}{4}$ torr iodine is $\frac{1}{3}$ that of the N_2 second positive (0-1) band at the same argon pressure and the same spectrometer slitwidth. Assuming no absorption by the excited iodine[17] and that only one upper excited state is involved, the gain from a laser operating at 342.6 nm will be $\frac{1}{5}$ that of the N_2 second positive laser operating at 357.7 nm. As can be seen from Fig. 1 the full width at half gain for the band will be $\simeq$ 2.5 nm.

An approximate value for the efficiency of a laser operating under the above conditions can be estimated as follows. Laser efficiency is the product of the argon pumping efficiency (50%), the fluorescence yield,[18] and the quantum efficiency (i.e., the ratio of the average stored energy in the noble gas to the energy emitted by the lasing molecule) which for argon and iodine at 340 nm is approximately 37%. To find the fluorescence yield the observed I_2 340 nm band emissions are integrated over time, and over wavelength from 300 to 350 nm; this value is 700 $\pm$ 180 volt-microseconds. The error limits arise from the Febetron pulse reproducibility and from the uncertainty in the integration over wavelength. A similar measurement and integration over the light emitted from the $N_2(C)$ under the same excitation conditions yields 87 $\pm$ 17 volt-microseconds. By multiplying the ratio of these two numbers by the 8.8% fluorescence yield for the $N_2(C)$ state determined from our Ar/N_2 modeling program,[3] we conclude that

the fluorescence yield for the Ar/I_2 system is 70 ± 24%. Total fluorescence efficiency for this band is then 13 ± 4%.

In an actual laser the efficiency could be larger than the above value since the stimulated emission may occur before the effects of quenching collisions can deplete the upper state population, an effect not accounted for in the measurements of fluorescence yield. We assume, of course, that due to collisional mixing in the high pressure gas, all of the upper state population can participate in the laser action. Furthermore, for these preliminary measurements, we have not made a systematic search for the optimum laser operating conditions. In a system as complicated as the argon/iodine, the optimization of all the properties, particularly the gain, efficiency and scaling behavior, will require a thorough understanding of the many formation and loss processes. In the immediate future we intend to investigate the kinetic processes of this highly promising laser candidate.

We would like to acknowledge the cooperation of Dr. L. D. Schearer who discussed with us some experimental results prior to their publication.

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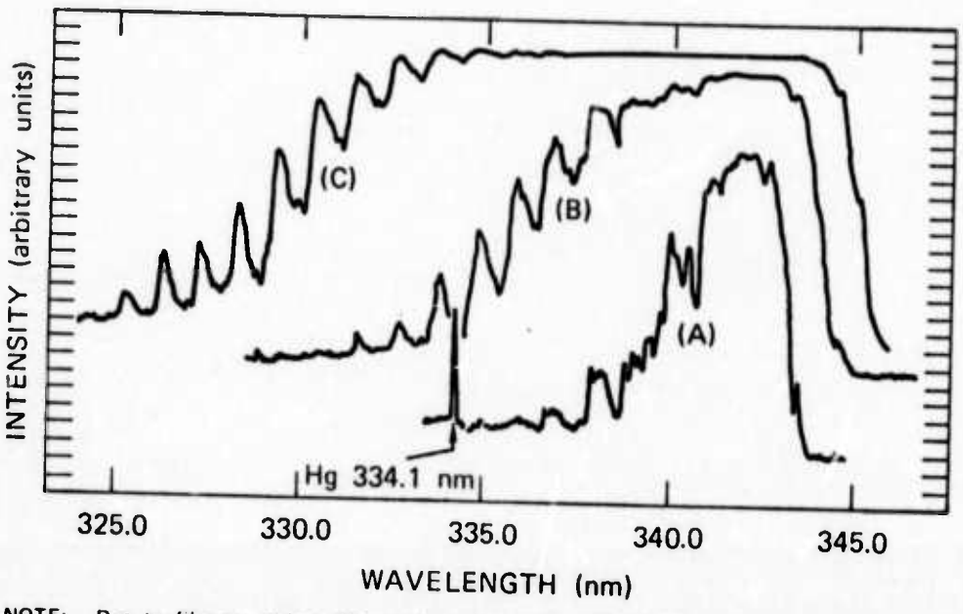
18. The fluorescence yield of a band is defined to be the ratio of the number of radiations emitted to the total number of excited rare gas species produced by the electrons. The excited argon density produced under these excitation conditions can be determined from the known Ar/N_2 fluorescence yield by measuring the N_2 second positive light flux from argon/nitrogen mixtures substituted into the reaction chamber.



NOTE: Due to film saturation effects the vertical scales are not linear and these data are presented for illustrative purposes only. Curve A is for two Febetron shots, Curve B is for 5, and Curve C is for 30. Also in the diagram is a single line from a mercury calibration source which is shown to illustrate tha spectrometer resolution. These spectra wara takan in second order.

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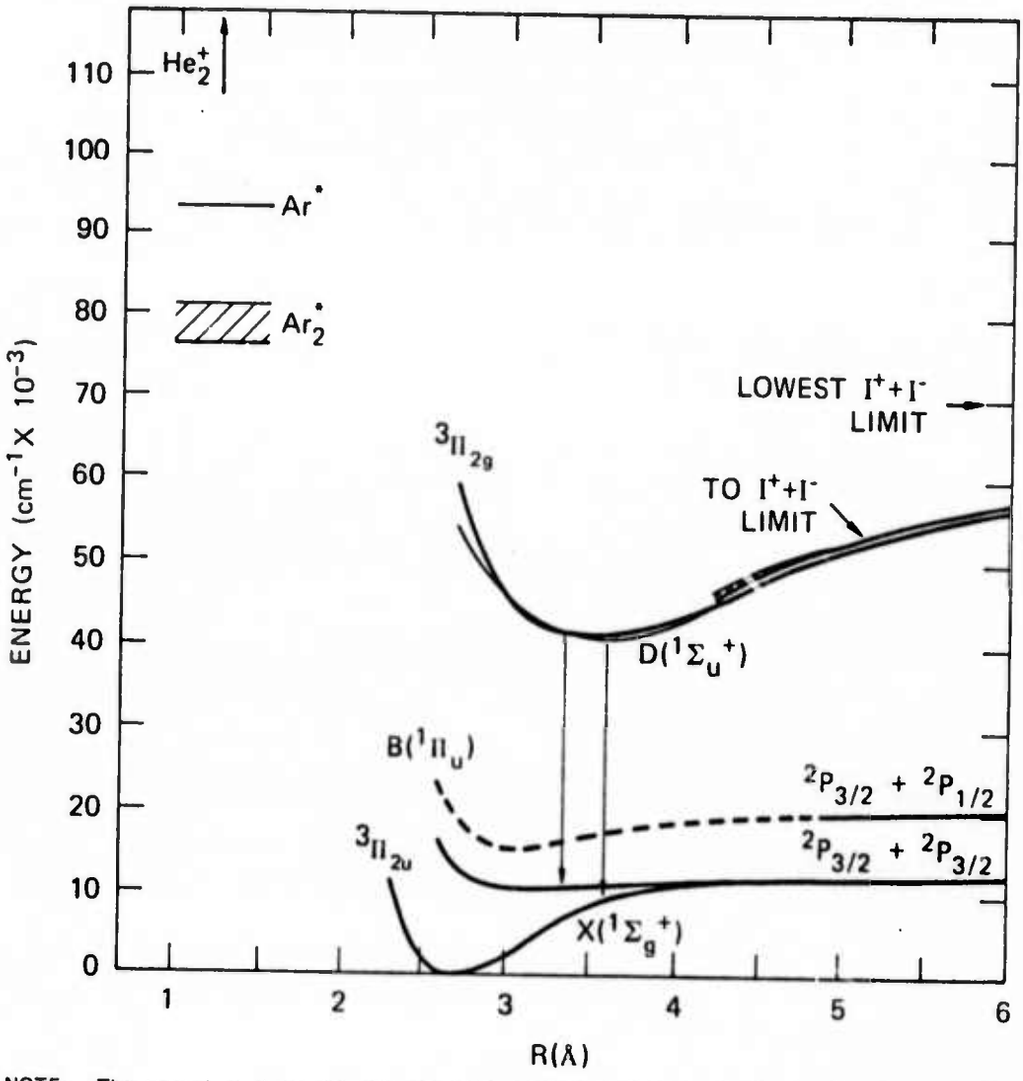
FIGURE 1 DENSITOMETER TRACES OF THE MOLECULAR IODINE EMISSION BAND NEAR 340 nm



NOTE: The transitions shown between the upper ionic curves and the lower covalent curves are believed to be responsible for the 340 nm radiation. For simplicity a large number of other potential curves are not included here. On the left are indicated the energies available from excited argon atoms and molecules present in the e-beam excited gas.

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FIGURE 2 RELEVANT POTENTIAL CURVES OF MOLECULAR IODINE, SELECTED FROM THOSE DESCRIBED BY MULLIKEN